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A PROCESS FOR PRODUCING POLYESTER RESIN

DECLARATION

Commissioner for Patents
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Sir:

I, Akiko KOJIMA, declare and say:

that I am thoroughly conversant in both the Japanese and English languages;

that I am presently engaged as a translator in these languages;

that the attached document represents a true English translation of the
Japanese Priority Application No. 2001-59316, filed March 2, 2001.

I further declare that all statements made herein of my own knowledge are true
and that all statements made on information and belief are believed to be true; and
further that these statements were made with the knowledge that willful false
statements and the like so made are punishable by fine or imprisonment, or both,
under Section 1001 of Title 18 of the United States Code, and that such willful false
statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 12th day of September, 2003.

Akiko Kojima

Akiko KOJIMA

(TRANSLATION)

JAPAN PATENT OFFICE

**This is to certify that the annexed is a true copy of the following
application as filed with this Office.**

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Applicant(s): Kansai Paint Co., Ltd.

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Kozo OIKAWA

**Commissioner,
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[Title of the Invention] A Process for Producing Polyester Resin

[Claims]

[Claim 1] A process for producing a polyester resin having a resin acid value
5 not higher than 120 mg KOH/g, a hydroxyl value not higher than 120 mg KOH/g
and a number-average molecular weight within a range of 2,000 to 30,000, the
process being characterized by concurrently reacting polyester whose chief
starting material is terephthalic acid and which has been recovered from waste
materials and regenerated, a polyhydric alcohol component and a polybasic
10 acid component, at such ratios that the regenerated polyester occupies 10 to
80% by weight, based on the total weight of said regenerated polyester, the
polyhydric alcohol component and the polybasic acid component.

[Claim 2] The process according to Claim 1, in which said polyester whose
chief starting materials is terephthalic acid and which has been recovered from
15 waste materials and regenerated is recycled polyethylene terephthalate.

[Claim 3] The process according to Claim 1 or 2, in which the chloroform-
insoluble component of the produced polyester resin is not more than 1.0%.

[Claim 4] A paint composition containing a polyester resin which is produced
by the process according to any one of Claims 1 to 3.

20 [Claim 5] The paint composition according to Claim 4, which is a powder
coating composition.

[Detailed Description of the Invention]

[0001]

[Technical field to which the invention belongs]

25 The present invention relates to a novel process for producing a
polyester resin using regenerated polyester which has been recovered from
waste materials such as disused PET (polyethylene terephthalate) bottles, and
also to paint compositions containing the polyester resin prepared by said
process.

30 [0002]

[Prior art and the problems]

Disposal of wastes has been recently causing serious problems, and
"3R's" (Recycle, Reduce and Reuse) are recommended. In particular, recycling

of disused PET bottles is being promoted, led by the Council for PET Bottle Recycling. Aided by the complete enactment of "Containers and Packagings Recycling Act" in April, 2000, the recovery ratio of disused PET bottles has been steadily increasing. Under the circumstances, it is essential to enlarge utility of the recovered and regenerated polyester (recycled polyester).

[0003]

For example, Japanese Patent Application Laid-Open No. 5(1993)-311114 and Japanese Patent Application Laid-Open No. 6(1994)-9923 disclose a process for producing the intended polyester resin, which comprises subjecting a polyhydric alcohol component and regenerated PET to depolymerization reaction in the presence of a catalyst to reduce the molecular weight of the PET, and adding a polybasic acid component to the system to carry out a polycondensation reaction. Those publications also teach that the resulting polyester resin is useful for both water-based and non-water-based coating compositions. However, there have been problems that the same process requires many hours for the depolymerization reaction of polyhydric alcohol component with regenerated PET, and the production costs are high compared to conventional processes which do not use regenerated polyester.

[0004]

Further, Japanese Patent Application Laid-Open No. 11(1999)-228733 discloses a process for synthesizing a polyester resin through an ester-interchange reaction of recycled polyester such as regenerated PET with a polyester oligomer having a molecular weight not higher than 5,000. However, this process is subject to a number of problems that it requires an extra step of advance synthesis of said polyester oligomer; its depolymerization step requires many hours because sufficient depolymerization of recycled polyester is difficult due to insufficient amount of alcohol component in the depolymerization system and also because it is a reaction between high molecular weight compounds; and the filtration residue is apt to form in large quantities.

[0005]

Furthermore, Japanese Patent Application Laid-Open No. 8(1996)-253596 discloses a production process of a binding resin for toners which comprises two steps of adding recycled polyester such as regenerated PET to

the reaction system to carry out a polymerization while depolymerizing the recycled polyester in the presence of alcohol and if necessary, water; and then of adding an acid component to carry out an addition reaction and increasing the molecular weight of the product. According to this process, the first step is
5 a reaction in the system in which a large excess (at least approximately twice) of hydroxyl groups to carboxyl groups are present, and depolymerization of recycled polyester predominantly takes place. Hence the polyester formed in the first step has a low molecular weight. In the second step, an acid component is blended to carry out the addition and polymerization reaction.

10 [0006]

An object of the present invention is to provide a process for producing, within a short time, a high molecular weight polyester resin free of any problem in respect of physical properties, which is transparent and substantially free of formation of foreign matters or filtration residues, using a
15 polyester resin recovered from waste materials and regenerated.

[0007]

[Means to solve the problems]

The present inventors have engaged in concentrative studies with the view to accomplish the above object, and now discovered that the object
20 can be met by concurrently reacting a polyester resin made chiefly from terephthalic acid, which has been recovered from waste materials and regenerated, with an alcoholic component and an acid component. The present invention has thus been completed.

[0008]

25 That is, the present invention provides a process for producing a polyester resin having a resin acid value not higher than 120 mg KOH/g, a hydroxyl value not higher than 120 mg KOH/g and a number-average molecular weight within a range of 2,000 to 30,000, the process being characterized by concurrently reacting polyester whose chief starting material is terephthalic acid
30 and which has been recovered from waste materials and regenerated, a polyhydric alcohol component and a polybasic acid component, at such ratios that the regenerated polyester occupies 10 to 80% by weight, based on the total weight of said regenerated polyester, polyhydric alcohol component and

polybasic acid component.

[0009]

The present invention furthermore provides paint compositions which contain the polyester resin produced by the above-described production
5 process.

Hereinafter the present invention is explained in further details.

[0010]

[Mode for carrying out the invention]

First, the production process of the present invention is explained in
10 detail.

Those polyester resins used in the process of the present invention, which are made chiefly from terephthalic acid and which have been recovered from waste materials and regenerated (hereafter the polyester resins may be abbreviated as "regenerated PES") include polyethylene terephthalate (e.g.,
15 PET bottles) which are recovered for recycling of sources, industrial waste polyethylene terephthalate, and polyester resins regenerated from the wastes occurring in the course of production of polyester products (film, fibers, etc.) made chiefly from terephthalic acid. In particular, the recycled polyethylene terephthalate and recycled polybutylene terephthalate are suitable. The
20 regenerated PES is normally used in the form of chips or crushed pieces.

[0011]

According to the process of the present invention, such regenerated PES, polyhydric alcohol component and polybasic acid component are concurrently reacted, if necessary in the presence of a depolymerization
25 catalyst.

[0012]

As said polyhydric alcohol component, polyhydric alcohols that are used as an alcohol component for constituting a polyester resin can be used, examples of which including dihydric alcohols such as ethylene glycol,
30 diethylene glycol, triethylene glycol, propylene glycol, 1,3-propanediol, 2-methyl-1,3-propanediol, 2,2-diethyl-1,3-propanediol, 2-butyl-2-diethyl-1,3-propanediol, neopentyl glycol, 1,4-butanediol, 1,5-pentanediol, 1,6-hexan diol and 1,4-dimethylolcyclohexane; and tri- and higher hydric alcohols such as

trimethylolpropane, trimethylolethane, glycerine, diglycerine, triglycerine, pentaerythritol, dipentaerythritol and sorbitol. Of those, glycerine, trimethylolpropane, ethylene glycol, neopentyl glycol and 1,4-dimethylolcyclohexane are preferred in view of their depolymerizing ability and

5 solubility.

[0013]

As the polybasic acid component described above, polybasic acids normally used as an acid component forming a polyester resin can be used, examples of which including dibasic acids such as phthalic anhydride,

10 isophthalic acid, terephthalic acid, tetrahydrophthalic anhydride, hexahydrophthalic anhydride, 1,4-cyclohexanedicarboxylic acid, 1,3-cyclohexanedicarboxylic acid, naphthalenedicarboxylic acid, 4,4'-biphenyldicarboxylic acid, diphenylmethane-4,4'-dicarboxylic acid, succinic acid, fumaric acid, adipic acid, sebacic acid and maleic anhydride; tri- and higher

15 polybasic acids such as trimellitic anhydride, pyromellitic anhydride, trimesic acid and methylcyclohexene-tricarboxylic acid; and lower alkyl esters of those dibasic acids or tri- and higher polybasic acids. Of those, dibasic acids or their lower alkyl esters are preferred.

[0014]

20 A compounding ratio of the regenerated PES, polyhydric alcohol component and polybasic acid component in the process of the present invention is such a quantitative ratio that the amount of the regenerated PES is within a range of, based on the total weight of said three components, 10 to 80% by weight, preferably 20 to 70% by weight, and that the polyester resin

25 obtained according to said process has a resin acid value not more than 120, preferably not more than 80, and a hydroxyl value not more than 120, preferably not more than 80.

[0015]

According to the process of the present invention, a

30 depolymerization reaction of the regenerated PES and an esterification reaction take place to form polyester resins. The term, "esterification reaction" as used herein includes not only an ordinary esterification reaction but also an "ester-interchange reaction". While the depolymerization of the regenerated PES

progresses in the absence of catalyst at reaction temperatures higher than the melting point of regenerated PES (at about 240°C and above), use of a depolymerization catalyst is preferred to carry out the depolymerization at temperatures lower than the melting point or to perform it more effectively.

5 [0016]

Examples of the depolymerization catalyst described above include monobutyltin hydroxide, dibutyltin oxide, monobutyltin-2-ethyl hexanoate, dibutyltin dilaurate, stannous oxide, tin acetate, zinc acetate, manganese acetate, cobalt acetate, calcium acetate, lead acetate, litharge, antimony trioxide, tetrabutyl titanate and tetraisopropyl titanate. A use amount of the depolymerization catalyst is preferred to be generally not more than 10 parts by weight, in particular, within a range of 0.001 to 5 parts by weight, per 100 parts by weight of the total of the regenerated PES, polyhydric alcohol and polybasic acid. Normally, the depolymerization catalyst can also serve as an esterification reaction (polymerization) catalyst.

15 [0017]

The process of the present invention comprises the reaction of the above-described regenerated PES, polyhydric alcohol component and polybasic acid component all in one system, in the optional presence of a depolymerization catalyst, in which the depolymerization reaction of the regenerated PES described above and the esterification reaction (including ester-interchange reaction) among the regenerated PES, polyhydric alcohol component and polybasic acid component are carried out concurrently to produce the intended polyester resin. This reaction is not limited so long as said depolymerization reaction and esterification reaction are conducted simultaneously. Whereas, generally the intended polyester resin can be produced by mixing the three components and reacting them under stirring at about 160 to about 270°C for about 2 to 10 hours.

25 [0018]

According to the process of the present invention, a characteristic of drastic reduction in reaction time is attained by the concurrent reaction.

30 [0019]

Conventionally, the intended polyester resin was synthesized by

conducting an alcoholysis reaction of the regenerated PES and, after finishing the decomposition reaction, adding acid monomers to the system to conduct a condensation reaction, for the purpose of an efficient decomposition of the regenerated PES. On the other hand, in the present invention, reactions such as an alcoholysis reaction of the regenerated PES and an ester-interchange reaction, and a condensation reaction of polyester both progress simultaneously and effectively, in the optional presence of a depolymerization catalyst.

[0020]

10 When the polyhydric alcohol component used in the reaction is in solid form, it is preferably heated in advance and melted.

[0021]

Thus produced polyester resin has a resin acid value not higher than 120 mg KOH/g, preferably not higher than 80 mg KOH/g; a hydroxyl value not higher than 120 mg KOH/g, preferably not higher than 80 mg KOH/g; and a number-average molecular weight within a range of 2,000 to 30,000, preferably 2,500 to 20,000. In particular, those having a resin acid value of 2.0 to 80 mg KOH/g, a hydroxyl value of 2.0 to 80 mg KOH/g and a number-average molecular weight of 2,500 to 10,000 are preferred. Furthermore, a chloroform-insoluble component of the produced polyester resin is preferably not more than 1.0%, that is, it is preferred that the regenerated PES has been sufficiently decomposed and the formed polyester has a uniform composition, from the viewpoint of a paint film appearance when the paint film is formed.

[0022]

25 After termination of the esterification reaction, the polyester resin is normally cooled and isolated from the reaction mixture as it is, or for improving its filterability and handling property, it may be diluted with an organic solvent and recovered. A kind of said organic solvent is subject to no limitation so long as it can dissolve the polyester resin.

30 [0023]

Those polyester resins produced according to the process of the present invention are useful as a resin binder in paint compositions, and when they have a glass transition temperature not lower than about 30°C, they can

be also used as a resin binder for powder coating compositions.

[0024]

When the polyester resins obtained according to the process of the present invention contain hydroxyl groups, they can be conveniently used as
5 resins for thermosetting paints, in combination with a curing agent which is reactive with hydroxyl groups, e.g., an amino resin such as a melamine resin, a polyisocyanate compound, an epoxy compound and the like. On the other hand, when the polyester resins contain carboxyl groups, they can be conveniently used as resins for thermosetting paints, in combination with a
10 curing agent which is reactive with carboxyl groups, such as an epoxy compound.

[0025]

For formulating a paint using the polyester resin obtained according to the process of the present invention, an organic pigment, an inorganic
15 pigment, a filler, a curing catalyst, a surface control agent (anti-cissing agent), a flow-and-leveling agent, an ultraviolet absorber, an ultraviolet stabilizer, an organic solvent and the like may be added if necessary, besides the polyester resin and curing agent described above. When the polyester resin has a glass transition temperature not lower than 30°C, it can be conveniently used as a
20 resin for powder coating compositions. For preparing such a powder coating composition, processes known per se for formulating powder coating compositions, e.g., processes by combining the polyester resin, the curing agent and, if necessary, other components, dry-blending them in a mixer, heat-melting, kneading, cooling, coarse crushing, finely pulverizing and classifying,
25 can be used.

[0026]

The paint in which the polyester resin obtained by the process of the present invention is used, can form a cured paint film upon being applied onto an object matter and baked. Baking conditions are not limited so long as the
30 applied paint can be cured thereunder. Normally, when the baking time is around 5 to 30 minutes, the baking temperature is preferably 120 to 210°C, and when the baking time is less than 5 minutes, the preferred baking temperature condition is that the maximum temperature of the object matter reaches 180 to

250°C.

[0027]

[Examples]

Hereinafter the present invention is more specifically explained with
5 reference to Examples in which parts and percentages are by weight.

[0028]

Example 1

A reactor equipped with a thermometer, a stirrer, a heater and a
rectification column was charged with 299 parts of neopentyl glycol and 41
10 parts of trimethylolpropane, which were heated to 130°C under stirring until the
whole system turned to a transparent liquid. Then, 1.0 part of monobutyltin
oxide, 448 parts of isophthalic acid and 547 parts of recycled polyethylene
terephthalate (regenerated PET) were charged and heated to 180°C. The
temperature of the content was then gradually raised from 180°C to 240°C over
15 a period of 3 hours, and the reaction was continued at 240°C for further 30
minutes. Then, the rectification column was replaced with a water separator,
and approximately 50 parts of xylene was added to the reaction mixture.
Xylene was poured also into the water separator, and condensed water was
removed through azeotropic distillation of water and xylene. Thirty minutes
20 after the addition of xylene, the acid value measurement was started and two
hours after the addition of xylene, heating was stopped upon confirming that the
acid value was less than 5.0. Removing the xylene under reduced pressure, a
polyester resin-1 was obtained as 100% heating residue. The production time
of said polyester resin from the initiation to suspension of the heating for
25 reaction was about 6.5 hours.

[0029]

Examples 2 to 4

Example 1 was repeated except that the blended composition was
varied for each run as shown in later given Table 1, to provide respective
30 polyester resins.

[0030]

Example 5

A reactor similar to the one used in Example 1 was charged with 299

parts of neopentyl glycol and 41 parts of trimethylolpropane, which was then heated to 130°C under stirring. When the system turned to a transparent liquid, 1.0 part of monobutyltin oxide, 647 parts of isophthalic acid and 547 parts of regenerated PET were added thereto, and heated to 180°C. The temperature of the system was then gradually raised from 180°C to 240°C over a period of 3 hours, and the reaction was continued for further 3 hours at 240°C while measuring the acid value. The heating was then stopped after confirming that the acid value was less than 53.0. Removing the water in the system by reduced pressure distillation, a polyester resin-5 was obtained as 100% heating residue. The production time of the polyester resin was about 7.0 hours, from the initiation to suspension of the heating for the reaction.

[0031]

Example 6

Example 1 was repeated except that said 1.0 part of monobutyltin oxide was replaced with 1.0 part of litharge, to produce a polyester resin-6. The production time of said polyester resin was about 7.5 hours, from the initiation to suspension of the heating for the reaction.

[0032]

Example 7

Example 1 was repeated except that the use of 1.0 part of monobutyltin oxide was omitted and that the gradual temperature rise from 180°C to 250°C was effected, after heating to 180°C, over a period of 3 hours. The subsequent reaction was continued at 250°C. Thus a polyester resin-7 was obtained. The production time of said polyester resin was about 8.5 hours, from the initiation to suspension of the heating for the reaction.

[0033]

Comparative Example 1

A reactor similar to the one used in Example 1 was charged with 299 parts of neopentyl glycol and 41 parts of trimethylolpropane, which were heated to 130°C under stirring. After the system was converted to a transparent liquid, 1.0 part of monobutyltin oxide and 547 parts of regenerated PET were charged and heated to 190°C. Thereafter, the temperature of the system was gradually raised, to an extent that the temperature at the upper portion of the rectification

column does not exceed 100°C. Thus, consuming about 6 hours until the content became transparent, the regenerated PET was reacted (including alcoholysis and ester-interchange reaction).

[0034]

5 Thereafter, the system in the reactor was cooled to 180°C, to which 448 parts of isophthalic acid was added and together gradually heated to 240°C over a period of 3 hours. The reaction was continued for further 30 minutes at 240°C, and the rectification column was replaced with a water separator. About 50 parts of xylene was added to the system, and xylene was fed also into the
10 water separator. The condensed water was removed through azeotropic distillation of water and xylene. After 1 hour from the addition of xylene, the acid value measurement was started, and 3.5 hours after the addition of xylene, the acid value was confirmed to be less than 5.0. Whereupon the heating was stopped and xylene was removed under reduced pressure, a polyester resin-8
15 whose 99.8% was the heating residue was provided. The production time of said polyester resin was about 16 hours, from the initiation to suspension of the heating for the reaction.

[0035]

Comparative Example 2

20 Example 5 was repeated except that the blended composition was changed as shown in later appearing Table 1 and that the reaction was continued for 5 hours after the temperature was raised to 230°C while measuring the acid value. Thus, a polyester resin-9 was obtained, in which white powder which presumably was attributable to sublimation of the acid
25 component remained, and a transparent and homogeneous polyester resin was not formed.

[0036]

 The acid value, hydroxyl value and number-average molecular weight of each of the resins obtained in Examples 1 to 7 and Comparative
30 Examples 1 and 2 were as shown in the following Table 1. The blended amounts as given in Table 1 are in terms of part by weight.

TABLE 1

	Example				
	1	2	3	4	5
Polyester resin	Polyester resin-1	Polyester resin-2	Polyester resin-3	Polyester resin-4	Polyester resin-5
Neopentyl glycol	299	299	158	110	299
1,6-Hexanediol				212	
Trimethylolpropane	41	41	41	41	41
Ethylene glycol		93			
Isophthalic acid	448	448	224	149	647
Terephthalic acid		249			
Adipic acid				263	
Regenerated PET	547	259	806	547	547
Monobutyltin oxide	1.0	1.0	1.0	1.0	1.0
Reaction time (hrs.)	6.5	7.5	6.5	6.5	7.0
Blended ratio of regenerated PET (%)	41	21	68	45	39
Resin acid value (mg KOH/g)	4.8	4.9	4.7	4.8	52.5
Hydroxyl value (mg KOH/g)	59	59	62	60	0
Glass transition point (°C)	55	54	60	-18	60
Number-average molecular weight	6600	6000	7000	6600	6100

[0038]

[Table 2]

TABLE 1 (continued)

	Example		Comparative Example	
	6	7	1	2
Polyester resin	Polyester resin-6	Polyester resin-7	Polyester resin-8	Polyester resin-9
Neopentyl glycol	299	299	299	299
1,6-Hexanediol				
Trimethylolpropane	41	41	41	41
Ethylene glycol				
Isophthalic acid	448	448	448	896
Terephthalic acid				
Adipic acid				
Regenerated PET	547	547	547	547
Monobutyltin oxide			1.0	1.0
Litharge	1.0			
Reaction time (hrs.)	7.5	8.5	16.0	9.0
Blended ratio of regenerated PET (%)	41	41	41	33
Resin acid value (mg KOH/g)	4.9	4.7	4.8	160
Hydroxyl value (mg KOH/g)	59	59	59	0
Glass transition point (°C)	55	55	55	60
Number-average molecular weight	6600	6600	6600	2500

[0039]

Example 8

One-hundred parts of the polyester resin-1 as obtained in Example 1, 30 parts of VESTAGON™ B-1530 (Hüls AG, Germany, ϵ -caprolactum-blocked triisocyanurate of isophoronediiisocyanate; solid content, 100%), 0.1 part of TAKENATE™ TK-1 (Takeda Chemical Industries, Ltd., an organotin-derived curing catalyst), 50 parts of TITANIX™ JR-605 (Tayca Corporation, a titanium white pigment), 1.0 part of POLYFLOW-S™ (Kyoisha Chemical Co., Ltd., a surface control agent) and 0.4 part of benzoin were dry-blended in Henschel mixer, heat-melted and kneaded in a twin screw extruder, cooled, coarsely crushed, finely pulverized with an atomizer and classified, to provide a thermosetting powder coating composition.

[0040]

Examples 9 to 13 and Comparative Example 3

Example 8 was repeated except that the blended composition was varied for each run as indicated in later appearing Table 2, to provide thermosetting powder coating compositions.

In said Table 2, the notes denote the following, respectively.

- (*1) EPIKOTE™ #1003: Shell Chemicals, Co., a bisphenol A-type epoxy resin
- (*2) CUREZOL™ 2E4MZ: Shikoku Chemicals Corporation, an imidazole-derived curing catalyst

[0041]

Example 14

One-hundred parts of the polyester resin-4, which was obtained in Example 4, were dissolved in 100 parts of a mixed solvent (remark 1), and the solution was mixed and stirred with 30 parts of VESTAGON™ B-1530, 0.1 part of TAKENATE™ TK-1, 50 parts of TITANIX™ JR-605, 1.0 part of POLYFLOW-S™ and 0.4 part of benzoin, using Atliter. After adjusting the viscosity of the resulting blend with said mixed solvent to 100 seconds (Ford Cup #4/25°C), the blend was filtered to provide a solvent type thermosetting paint.

(Remark 1) mixed solvent: SWASOL™ 1500 (Cosmo Oil Co., Ltd., a high-boiling hydrocarbon solvent) / cyclohexanone = 50/50 (weight ratio) solvent mixture

[0042]

Preparation of test panels

Each of the thermosetting powder coating compositions as prepared in Examples 8 to 13 and Comparative Example 3 described above were applied
5 onto a zinc phosphate-treated sheet steel by electrostatic powder coating, to a cured coating thickness of about 60 μm , and baked at 180°C for 20 minutes to provide test panels.

[0043]

Also the solvent type thermosetting paint, which was prepared in
10 Example 14 described above, was applied onto a zinc phosphate-treated sheet steel with a bar coater, to a cured film thickness of about 60 μm , and baked at 180°C for 20 minutes to provide a test panel.

[0044]

Those test panels described above were tested in respect of the
15 appearance of the paint film, 60° mirror surface gloss and impact resistance and given Erichsen test, according to the following test methods. The results were as given in the later appearing Table 2.

[0045]

Test methods

20 Paint film appearance:

The finish of the painted surface of each test panel was visually observed and evaluated according to the following standard;

O: good Δ : inferior x: poor.

[0046]

25 60° Mirror surface gloss:

Following JIS K-5400 7.6 (1990) 60° Mirror Surface Gloss, the gloss level of each paint film was determined by measuring its reflectivity when both incident angle and acceptance angle were 60°, and by expressing the result in percent to the gloss (100) of the standard mirror surface.

30 [0047]

Impact resistance:

Each test panel was allowed to stand in a 20°C atmosphere for about 24 hours, and then in the same 20°C atmosphere, it was placed on the

cradle of DuPont Impact Tester, with its painted surface on the top. A shock mould with semi-spherical front of about 6.35 mm in radius was placed on the painted surface, and a 500 g weight was dropped onto the hitting core. The impact resistance of the painted surface was judged by the height (cm) from which said weight was dropped, when the depression formed in the test specimen by the impact and the peripheral portion thereof started to crack. When no crack was caused by the weight dropping from the height of 50 cm, the impact resistance was indicated as 50<.

[0048]

10 Erichsen test:

Using Erichsen tester, each test panel was pushed from the side opposite to the painted surface, and the height (mm) of the pushed out portion at which cracks appeared in the paint film was recorded. When no crack was formed in the paint film at the pushed out height of 7 mm, the result was

15 indicated as 7<.

[0049]

[Table 3]

TABLE 2

		Example												Comparative Example
		8	9	10	11	12	13	14	Example					
		Kind (Example No.)		1	2	3	5	6	7	4	Comparative Example 1			
Polyester resin	Amount (part)	100	100	100	70	100	100	100	100	100	100	100	100	
	VESTAGON™ B-1530	30	30	30			30	30	30	30	30	30	30	
EPIKOTE™ #1003 (*1)					30									
TAKENATE™ TK-1		0.1	0.1	0.1		0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	
CUREZOL™ 2E4MZ (*2)					0.3									
TITANIX™ JR-605		50	50	50	50	50	50	50	50	50	50	50	50	
POLYFLOW-S™		1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	
Benzoin		0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	
Test Result	Appearance of paint film	○	○	○	○	○	○	○	○	○	○	○	○	
	60° Mirror surface gloss	96	95	94	92	94	93	94	93	95	95	95	95	
	Impact resistance	50<	50<	50<	50<	50<	50<	50<	50<	50<	50<	50<	50<	
	Erichsen test	7<	7<	7<	7<	7<	7<	7<	7<	7<	7<	7<	7<	

[0050]

[Effect of the invention]

According to the process of the present invention, a polyester resin which is transparent and substantially free of occurrence of foreign matters or
5 filtration residue as well as of any problem in physical property can be produced within short time, using a polyester resin recovered from waste materials. The process of the present invention enables utilization of disused PET bottles and the like for recycling, to utilize waste matters.

The polyester resin obtained according to the process of the present
10 invention, furthermore, can be conveniently used as a resin for thermosetting paint, in combination with a curing agent.

[Document Name] Abstract

[Abstract]

[Subject]

A polyester resin which is transparent and substantially free of occurrence of foreign matters or filtration residue as well as of any problem in physical property is produced within short time, using a polyester resin recovered from waste materials and regenerated.

[Means for Solution]

A process for producing a polyester resin having a resin acid value not higher than 120 mg KOH/g, a hydroxyl value not higher than 120 mg KOH/g and a number-average molecular weight within a range of 2,000 to 30,000, the process being characterized by concurrently reacting polyester whose chief starting material is terephthalic acid and which has been recovered from waste materials and regenerated, a polyhydric alcohol component and a polybasic acid component, at such ratios that the regenerated polyester occupies 10 to 80% by weight, based on the total weight of said regenerated polyester, polyhydric alcohol component and polybasic acid component.

[Selected Drawing]

None